

We would like to thank the reviewer for the helpful comments and suggestions. We answer each of them here after, with the original comment in blue and our response in black. We added the modifications done in the revised version in italics.

- [Referee #2] I suggest altering the title to something like “Improved method for mobile characterisation of $^{13}\text{CH}_4$ source signatures and its application in Germany.” The current title sells short of a major contribution of the paper.
- [Hoheisel et al.] Thank you for pointing this out. We have changed the title.
- Detailed comments follow:
- [Referee #2] 1. P 1, In 3: Remove “Therefore”; This sentence doesn’t logically follow from the previous sentence. It is detail in addition to the previous sentence.
- [Hoheisel et al.] Yes, corrected.
- [Referee #2] 2. P 1, In 5: Explain gas matrix or replace with less jargon.
- [Hoheisel et al.] Ok, we have changed it.
“To achieve precise results a CRDS analyser, ..., was characterised especially with regard to cross sensitivities of composition differences of the gas matrix in air samples or calibration tanks”.
- [Referee #2] 3. P 1, In 7: Abundant in many natural gases, but not all. Dry gas regions can contain only very small traces of ethane.
- [Hoheisel et al.] Yes, we have changed it.
“ C_2H_6 is typically abundant in natural gases”.
- [Referee #2] 4. P 1, In 16: A 2.8 per mil difference begs the question whether the above mentioned up to 3 per mil ethane bias could have played a role here.
- [Hoheisel et al.] In the 1990s Levin et al. (1999) measured $\delta^{13}\text{CH}_4$ with a mass spectrometer after separation of CH_4 from other components of air and so there is no cross-sensitivity with C_2H_6 .
- [Referee #2] 5. Switching between “ CH_4 ” and “methane” throughout the MS. Check for consistency.
- [Hoheisel et al.] Thanks, “methane” has been replaced in the manuscript by “ CH_4 ”.
- [Referee #2] 6. P 1, In 20: Use original data references instead of reviews, e.g., <https://agupubs.onlinelibrary.wiley.com/doi/full/10.1029/2009GL039780>
- [Hoheisel et al.] Yes, corrected.
- [Referee #2] 7. P 2, In 1: You probably mean biomass burning.
- [Hoheisel et al.] Thanks, corrected.
- [Referee #2] 8. P 2, In 2: More accurate to say “e.g. a sub-category of fossil fuel extraction”. Some gases extracted by the fossil fuel industry is biogenic.
- [Hoheisel et al.] Clarified.
- [Referee #2] 9. P 2, In 7: Do you mean to say that the isotopic signal was used to diagnose methane emission reductions? Not clear as written.
- [Hoheisel et al.] Agreed, we have clarified it.
- [Referee #2] 10. P 2, In 13: What do you mean with “all of these seasonal variations”?
- [Hoheisel et al.] Yes, this was unclear. We wanted to write “*seasonal variations*”.
- [Referee #2] 11. P 2, In 26: Briefly describe the importance/use of a storage tube (first mention).
- [Hoheisel et al.] Done.
- [Referee #2] 12. P 2, In 26: Nine facilities in 21 campaigns? Do you mean 21 measurement days?
- [Hoheisel et al.] Yes, we have changed it to “*21 mobile measurement days*”.

- [Referee #2] 13. P 3, In 15: Perhaps that's explained later, but how do you get so close to the source that you're able to sample between 50 and 90% CH₄?
- [Hoheisel et al.] We take gas samples for example directly from the gas collecting system of the landfill and the WWTP. We now have clarified it and changed the phrase.
"Gas samples taken directly from different installations (e.g. natural gas pipelines, biogas plants, gas collecting systems of landfills and wastewater treatment plants) need to be diluted before the measurement with the CRDS analyser, because such samples usually consist of between 50 and 90% CH₄."
- [Referee #2] 14. P 3, In 17: Specify the composition of synthetic air.
- [Hoheisel et al.] We now have added the composition (20.5±0.5% O₂ in N₂).
- [Referee #2] 15. P 3, In 33: Do you mean a 20-25 sec lag time between air sampling at inlet and fully arriving in the cavity? Sounds like it, but not fully clear.
- [Hoheisel et al.] Yes, we clarified it.
"Due to the length of the intake line, the volume of the cavity, and a flow rate of 160ml/min the time lag between air sampling at inlet and measurement in the cavity of the CRDS analyser is 20 to 25sec."
- [Referee #2] 16. P 4, In 11: What do you mean with representations have the same width? They don't. It's a bit confusing. The next sentence already states that both peaks represent the same emission plume.
- [Hoheisel et al.] What we meant is that when measuring a CH₄ plume first without and subsequently with the AirCore the two measured CH₄ peaks have the same height, but since we measured with a slower flow in monitoring mode not the same width. For better comparison we stretched the peak measured without the AirCore in x direction, to make it easier to compare the measurements done with and without the AirCore and to note that both peaks have the same shape and height. We also changed the text to:
"For comparison the peak measured in 'monitoring mode' (blue dots/line on the left side) is stretched by a factor of 12.5 in x-direction (blue line on the right side) so that the peak measured with the AirCore and the stretched one measured without it have the same width. The peak measured in 'replay mode' precisely corresponds to the stretched one measured in 'monitoring mode', because both peaks reproduce the same emission plume."
- [Referee #2] 17. P 4, In 13: Any hypotheses why this may be the case? This seems to be an important observation.
- [Hoheisel et al.] As described above we measured the air stored in the AirCore directly after we measured a CH₄ peak in monitoring mode. So the storage time is relatively short and we expect that the peaks measured with and without the AirCore have the same shape and the same height.
- [Referee #2] 18. Table 1: Is the unit for ethane sensitivity to water (ppm) a typo? Units of ppb would make more sense. For reference, atmospheric ethane background concentrations are in the order of < 1 ppb.
- [Hoheisel et al.] No, the unit ppm is correct.
- [Referee #2] 19. P 5, In 5: Natural gases on a continuum between 0 to 40% ethane have been measured. See Sherwood et al., 2017 (already in your refs). Hence, the importance of ethane correction varies: very important for wet gas basins (meaning lots of associated gas) and less important for very dry gas regions (mature thermogenic dry gas).
- [Hoheisel et al.] Thank you for pointing this out. In this study we measured natural gas samples from the natural gas network in Heidelberg. Therefore, we wanted to give an overview of C₂H₆ contents of natural gases in the pipeline network in Germany. We have now added:
"As typical natural gases in the pipeline network in Germany contain..."

- [Referee #2]
[Hoheisel et al.] 20. P 5, In 10: where does the 3 per mil value come from?
The value of 3‰, is the bias $\delta^{13}\text{CH}_4$ has in presence of a C_2H_6 to CH_4 ration of 0.073, which was the highest value we measured for our natural gas samples. To make the text more understandable, we changed it.
“A correction is necessary because for typical C_2H_6 to CH_4 ratios between 0.027 and 0.073 measured for our natural gas samples, $\delta^{13}\text{CH}_4$ shows a bias between 1 and 3‰ to more enriched values. We must also keep in mind that similar shifts in, $\delta^{13}\text{CH}_4$ to less enriched values can occur when using a calibration cylinder which contains C_2H_6 .”
- [Referee #2]
[Hoheisel et al.] 21. P 7, In 3: What is the sign of the drift? Is the drift due to fractionation in the bag or due to leakage of background air into the bag?
The sign is + and the major cause for the drift is fractionation by the leakage of sample air out of the bag. So for a better understanding we added the phrase “to more enriched values” and the following sentence.
“The drift occurs especially due to fractionation by diffusion of air through the sample bag”.
- [Referee #2]
[Hoheisel et al.] 22. P 7, In 28: What are simulated data?
How we simulated data is explained a few lines below. For better understanding we changes this section:
“Similar to the method described by Wehr and Saleska (2017) for CO_2 and $\delta^{13}\text{CO}_2$, we create several typical emission plume crossings. We generated synthetic CH_4 peaks using a background concentration of 1.95 ppm CH_4 and a Gaussian curve with 10-280 equidistant data points every 3.7 s and an enhancement of 100-10000 ppb. The corresponding $\delta^{13}\text{CH}_4$ values were calculated with CH_4 source signatures between -35‰ and -65‰ and a background of -48‰. To reproduce the statistical uncertainties of a real measurement, we add a normally distributed scattering around zero to the synthetic CH_4 concentrations and the corresponding isotope ratios. The standard deviation of the normal distributed scattering depends on the CH_4 concentrations and was chosen as the Allan standard deviation measured for raw data of the analyser. However, when simulating possible improved analysers, we reduced the scattering by a factor 2 to 10. Such sets of data were generated 5000 times for each condition. To study the influence of the averaging time, we calculate mean data sets with varying averaging periods (up to 1min). For each dataset the $\delta^{13}\text{CH}_4$ source signature was calculated with the Miller-Tans and the Keeling method using the York or the OLS fit.”
- [Referee #2]
[Hoheisel et al.] 23. P 10, In 4: I'm confused. Here it says no significant seasonal cycle has been observed. In the next paragraph, it says the values are more depleted in winter than in summer. Are there enough samples to determine this correctly?
We changed this paragraph.
- [Referee #2]
[Hoheisel et al.] 24. P 10, In 13: What are the uncertainties for the ethane-to-methane ratios?
The uncertainties for the ethane-to-methane ratios is in both cases 0.01. We now have included it.
- [Referee #2]
[Hoheisel et al.] 25. P 12, In 2: Why were the plumes expected to be smaller than on the other farm?
Both farms have a different number of dairy cows. We now added the comment “due to lower animal number”.
- [Referee #2]
[Hoheisel et al.] 26. P 12, In 11: Why are seasonal differences for the biogas plant expected?
Thanks, we have removed “seasonal”.
- [Referee #2]
[Hoheisel et al.] 27. P 12, par. 1 & 3: Is C3/C4 diet information available for the Ladenburg and Kleve farms?
No, unfortunately not.

- [Referee #2] 28. P 14, In 11: How small the fluxes were actually depends on the size of the facility (is it possible to detect all plumes at once?) and the wind conditions in addition to the measured methane concentrations.
- [Hoheisel et al.] The facility is in a small forest and so in difficult terrain. It is unlikely that we detect all plumes at once. So we weakened our statement.
- [Referee #2] 29. P 14, In 20: Did your measurements include incomplete combustion from the compressor turbines? This could be detected by the associated CO₂ or CO signal. It is still an open question whether high heat leads to isotopic fractionation of the source.
- [Hoheisel et al.] To determine the origin of a CH₄ plume measured around the natural gas facility between Hähnlein und Gernsheim is difficult, because this facility contains a natural gas storage and several compressor stations even operated by different gas providers. At this site further work and more measuring campaigns are planned to receive more detailed results.
- [Referee #2] 30. P 14, In 25: Do you use “open” and “in service” interchangeably? Not clear. If yes, it’s not a surprise that a mine currently in service produces more emissions than a closed mine.
- [Hoheisel et al.] Yes, we changed “open” to “in service”.
- [Referee #2] 31. P 14, In 30: The Bottrop mine shaft measurements do not match coal bed gas samples except for the closed mine with a large error bar and only one AirCore measurement.
- [Hoheisel et al.] Done.
- [Referee #2] 32. Conclusions: In the first paragraph, it’s important to highlight again that these results (including the ethane bias) are specific to the CRDS instrument used.
- [Hoheisel et al.] OK, we have added the following phrase to highlight your recommendation.
“characterisation of each individual analyser”